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APPLICANT: Mari Ichimura et al. GROUP ART UNIT: 1774

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TITLE: ORGANIC ELECTROLUMINESCENT DEVICES,
AMINOSTYLNAPHTHALENE COMPOUNDS AND SYNTHESIS
INTERMEDIATES THEREOF, AND PRODUCTION PROCESSES
OF THE SAME

Hon. Commissioner of Patents and Trademarks,
Washington, D.C. 20231

S I R:

CERTIFIED TRANSLATION

I, Masaaki Iwami of 3-22, Asagaya-minami 1-chome,
Suginami-ku, Tokyo, Japan, am an experienced translator of
the Japanese language into the English language and I hereby
certify that the attached comprises an accurate translation
into English of Japanese Patent Application No. 2003-079768
filed March 24, 2003.

I hereby declare that all statements made herein of my
own knowledge are true and that all statements made on
information and belief are believed to be true; and further
that these statements were made with the knowledge that willful
false statements and the like so made are punishable by fine
or imprisonment or both, under Section 1001 of Title 18 of
the United States Code and that such willful false statements
may jeopardize the validity of the application or any patent
issued thereon.

February 26, 2007

Date



Masaaki IWAMI

[Name of Document] Specification

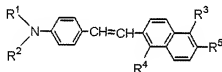
[Title of the Invention] Organic Electroluminescent
Devices, Aminostyrylnaphthalene Compounds and Synthesis
Intermediates thereof, and Production Processes of the
Same

[What is Claimed is]

[Claim 1] An organic electroluminescent device
comprising an organic layer having a light-emitting area
and arranged between an anode and a cathode, wherein at
least a part of said organic layer includes at least one
aminostyrylnaphthalene compound represented by the
following Formula [I], [II] or [III]:

[Chemical Formula 1]

Formula [I]



in the Formula [I], where

R¹ and R² may be the same or different and each
independently represents a phenyl group represented by
the following Formula (1),

[Chemical Formula 2]

Formula (1)



in the Formula (1), where R⁶, R⁷, R⁸, R⁹ and R¹⁰ may be the same or different, at least one of R⁶ to R¹⁰ represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom with a proviso that, said two adjacent ones of R⁶ to R¹⁰ may be fused together to form a ring, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R⁶ to R¹⁰ are each a hydrogen atom,

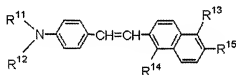
R³ and R⁴ may be the same or different, at least one of R³ and R⁴ represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R⁵ represents a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or unsubstituted alkoxyl group, a substituted or

unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group;

[Chemical Formula 3]

Formula [II]



in the Formula [II], where

R^{11} and R^{12} may be the same or different and each independently represents a naphthyl group represented by the following Formula (2),

[Chemical Formula 4]

Formula (2)



or

in the Formula (2), where R^{16} , R^{17} , R^{18} , R^{19} , R^{20} , R^{21} and R^{22} may be the same or different, at least one of R^{16} to R^{22} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or

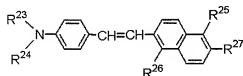
unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^{16} to R^{22} are each a hydrogen atom,

R^{13} and R^{14} may be the same or different, at least one of R^{13} and R^{14} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R^{15} represents a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or unsubstituted alkoxyl group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group;

[Chemical Formula 5]

Formula [III]

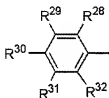


in the Formula [III], where

R^{23} is a phenyl group represented by the following
Formula (3),

[Chemical Formula 6]

Formula (3)

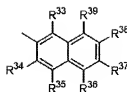
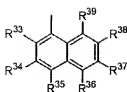


in the Formula (3), where R^{28} , R^{29} , R^{30} , R^{31} and R^{32} may be the same or different, at least one of R^{28} to R^{31} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom with a proviso that, said two adjacent ones of R^{28} to R^{31} may be fused together to form a ring, a hydrocarbyloxy group having at least one carbon atom, a hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^{28} to R^{31} are each a hydrogen atom,

R^{24} represents a naphthyl group represented by the following Formula (4),

[Chemical Formula 7]

Formula (4)



or

in the Formula (4), where R^{33} , R^{34} , R^{35} , R^{36} , R^{37} , R^{38} and R^{39} may be the same or different, at least one of R^{33} to R^{39} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^{33} to R^{39} are each a hydrogen atom,

R^{25} and R^{26} may be the same or different, at least one of R^{25} and R^{26} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R^{27} represents a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or

unsubstituted alkoxy group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group.

[Claim 2] The organic electroluminescent device according to claim 1, wherein said organic layer is in a form of an organic layer structure including a hole transport layer and an electron transport layer stacked one over the other, and at least said electron transport layer in said organic layer includes at least said one aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III].

[Claim 3] The organic electroluminescent device according to claim 1, wherein said organic layer is in a form of an organic layer structure including a hole transport layer and an electron transport layer stacked one over the other, and at least said hole transport layer in said organic layer includes at least said one aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III].

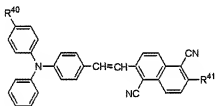
[Claim 4] The organic electroluminescent device according to claim 1, wherein said organic layer is in a form of an organic layer structure including a hole transport layer, a luminescent layer and an electron

transport layer stacked one over another, and at least said luminescent layer in said organic layer includes at least said one aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III].

[Claim 5] The organic electroluminescent device according to claim 1, wherein said aminostyrylnaphthalene compound is represented by the following Formula (5), (6), (7), (8), (9), (10), (11), (12), (13), (14), (15), (16) or (17):

[Chemical Formula 8]

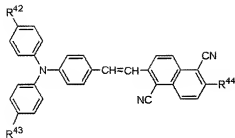
Formula (5)



in the Formula (5), where R^{40} represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R^{41} has the same meaning as R^5 ;

[Chemical Formula 9]

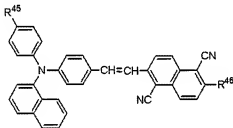
Formula (6)



in the Formula (6), where R⁴² and R⁴³ may be the same or different and each independently represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁴⁴ has the same meaning as R⁵;

[Chemical Formula 10]

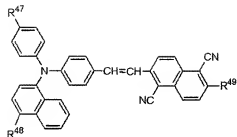
Formula (7)



in the Formula (7), where R⁴⁵ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁴⁶ has the same meaning as R²⁷;

[Chemical Formula 11]

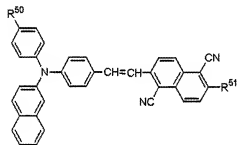
Formula (8)



in the Formula (8), where R⁴⁷ and R⁴⁸ may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁴⁹ has the same meaning as R²⁷;

[Chemical Formula 12]

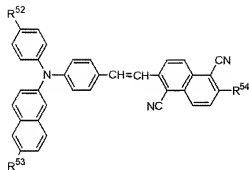
Formula (9)



in the Formula (9), where R⁵⁰ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁵¹ has the same meaning as R²⁷;

[Chemical Formula 13]

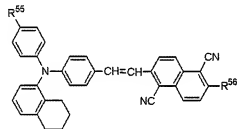
Formula (10)



in the Formula (10), where R⁵² and R⁵³ may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁵⁴ has the same meaning as R²⁷;

[Chemical Formula 14]

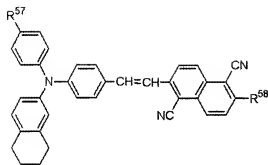
Formula (11)



in the Formula (11), where R⁵⁵ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁵⁶ has the same meaning as R⁵;

[Chemical Formula 15]

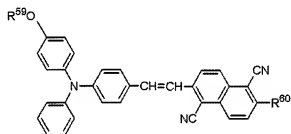
Formula (12)



in the Formula (12), where R⁵⁷ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁵⁸ has the same meaning as R⁵;

[Chemical Formula 16]

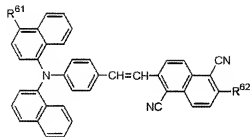
Formula (13)



in the Formula (13), where R⁵⁹ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁶⁰ has the same meaning as R⁵;

[Chemical Formula 17]

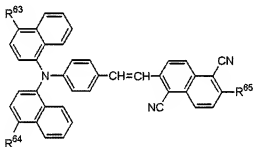
Formula (14)



in the Formula (14), where R⁶¹ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁶² has the same meaning as R¹⁵;

[Chemical Formula 18]

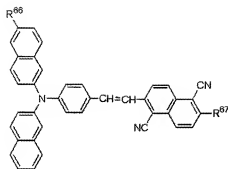
Formula (15)



in the Formula (15), where R⁶³ and R⁶⁴ may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁶⁵ has the same meaning as R¹⁵;

[Chemical Formula 19]

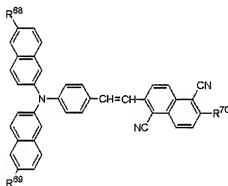
Formula (16)



in the Formula (16), where R⁶⁶ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁶⁷ has the same meaning as R¹⁵;

[Chemical Formula 20]

Formula (17)

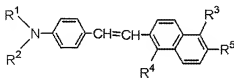


in the Formula (17), where R⁶⁸ and R⁶⁹ may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁷⁰ has the same meaning as R¹⁵.

[Claim 6] An aminostyrylnaphthalene compound, which is represented by the following Formula [I], [II] or [III]:

[Chemical Formula 21]

Formula [I]



in the Formula [I], where

R^1 and R^2 may be the same or different and each independently represents a phenyl group represented by the following Formula (1),

[Chemical Formula 22]

Formula (1)



in the Formula (1), where R^6 , R^7 , R^8 , R^9 and R^{10} may be the same or different, at least one of R^6 to R^{10} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom with a proviso that, said two adjacent ones of R^6 to R^{10} may be fused together to form a ring, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of

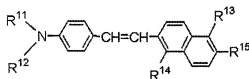
R⁶ to R¹⁰ are each a hydrogen atom,

R³ and R⁴ may be the same or different, one of R³ and R⁴ represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R⁵ represents a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group;

[Chemical Formula 23]

Formula [II]

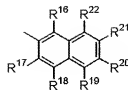
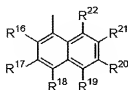


in the Formula [II], where

R¹¹ and R¹² may be the same or different and each independently represents a naphthyl group represented by the following Formula (2),

[Chemical Formula 24]

Formula (2)



or

in the Formula (2), where R¹⁶, R¹⁷, R¹⁸, R¹⁹, R²⁰, R²¹ and R²² may be the same or different, at least one of R¹⁶ to R²² represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R¹⁶ to R²² are each a hydrogen atom,

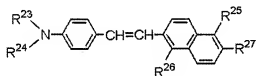
R¹³ and R¹⁴ may be the same or different, one of R¹³ and R¹⁴ represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R¹⁵ represents a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or

unsubstituted alkoxy group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group;

[Chemical Formula 25]

Formula [III]



in the Formula [III], where

R^{23} is a phenyl group represented by the following

Formula (3),

[Chemical Formula 26]

Formula (3)



in the Formula (3), where R^{28} , R^{29} , R^{30} , R^{31} and R^{32} may be the same or different, at least one of R^{28} to R^{31} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom with a proviso that, said two adjacent ones of R^{26} to R^{31} may be fused together to form a ring, a hydrocarbyloxy group having at least one carbon atom, a hydrocarbylamino group

having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^{28} to R^{31} are each a hydrogen atom, and

R^{24} represents a naphthyl group represented by the following Formula (4),

[Chemical Formula 27]

Formula (4)



or

in the Formula (4), where R^{33} , R^{34} , R^{35} , R^{36} , R^{37} , R^{38} and R^{39} may be the same or different, at least one of R^{33} to R^{39} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^{33} to R^{39} are each a hydrogen atom,

R^{25} and R^{26} may be the same or different, at least one of R^{25} and R^{26} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a

halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

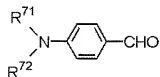
R^{27} represents a hydrogen atom, a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or unsubstituted alkoxyl group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group.

[Claim 7] The aminostyrylnaphthalene compound according to claim 6, which is represented by the Formula (5), (6), (7), (8), (9), (10), (11), (12), (13), (14), (15), (16) or (17) as defined in claim 5.

[Claim 8] A process for the production of an aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III] as defined in claim 6, which comprises subjecting a 4-(N,N-diarylamino)benzaldehyde represented by the following Formula [IV] and at least one of a phosphonate ester represented by the following Formula [V] and a phosphonium represented by the following Formula [VI] to condensation:

[Chemical Formula 28]

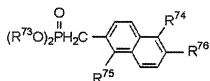
Formula [IV]



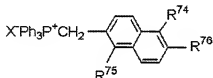
in the Formula [IV], where R^{71} and R^{72} each independently represents an aryl group corresponding to R^1 , R^2 , R^{11} , R^{12} , R^{23} or R^{24} as defined above;

[Chemical Formula 29]

Formula [V]



Formula [VI]



in the Formulas [V] and [VI], where R^{73} represents a hydrocarbon group, R^{74} and R^{75} each independently represents a group corresponding to R^3 , R^4 , R^{13} , R^{14} , R^{25} or R^{26} as defined above, R^{76} represents a group corresponding to R^5 , R^{15} or R^{27} as defined above, and X represents a halogen atom.

[Claim 9] The process according to claim 8, wherein said condensation is conducted by the Wittig-Horner

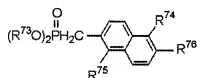
reaction or the Wittig reaction, at least one of said phosphonate ester and said phosphonium is treated with a base in a solvent to form carbanions, and said carbanions and said 4-(N,N-diarylamino)benzaldehyde are subjected to condensation.

[Claim 10] The process according to claim 8, wherein an aminostyrylnaphthalene compound represented by the Formula (5), (6), (7), (8), (9), (10), (11), (12), (13), (14), (15), (16) or (17) as defined in claim 5 is obtained.

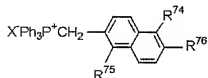
[Claim 11] The phosphonate ester or phosphonium, which is represented by the following formula [V] or [VI]:

[Chemical Formula 30]

Formula [V]



Formula [VI]



in the Formulas [V] and [VI], where R^{73} represents a hydrocarbon group, R^{74} and R^{75} each independently represents a group corresponding to R^3 , R^4 , R^{13} , R^{14} , R^{25} or

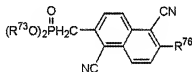
R^{26} , R^{76} represents a group corresponding to R^5 , R^{15} or R^{27} ,
and X represents a halogen atom.

[Claim 12] The phosphonate ester according to claim 11,
wherein R^{73} represents a saturated hydrocarbon group
having 1 to 4 carbon atoms.

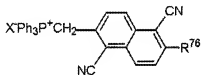
[Claim 13] The phosphonate ester or phosphonium
according to claim 11, which is represented by the
following Formulas (18) and (19):

[Chemical Formula 31]

Formula (18)



Formula (19)



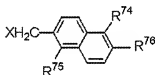
in the Formulas (18) and (19), where R^{73} , R^{76} and X
have the same meanings as defined above.

[Claim 14] A process for the production of a
phosphonate ester or phosphonium represented by the
formula [V] or [VI] as defined in claim 11, which
comprises reacting a halogenated aryl compound
represented by the following formula [VII] with a

trialkyl phosphite represented by the following formula
[VIII] or triphenylphosphine (PPh₃):

[Chemical Formula 32]

Formula [VII]



Formula [VIII]

P(OR⁷⁷)₃

in the Formula [VII] and [VIII], where R⁷⁴, R⁷⁵,
R⁷⁶ and X have the same meanings as above, and R⁷⁷
represents a hydrocarbon group.

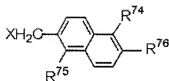
[Claim 15] The process according to claim 14, wherein
R⁷⁷ represents a saturated hydrocarbon group having 1 to 4
carbon atoms.

[Claim 16] The process according to claim 14, wherein
a phosphonate ester or phosphonium represented by the
formula (18) or (19) as defined in claim 13 is obtained.

[Claim 17] A halogenated aryl compound, which is
represented by the following Formula [VII]:

[Chemical Formula 33]

Formula [VII]

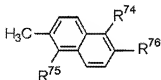


in the Formula [VII], where R^{74} and R^{75} may be the same or different, at least one of R^{74} and R^{75} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom; R^{76} represents a hydrogen atom, a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group; and X represents a halogen atom.

[Claim 18] A process for the production of a halogenated aryl compound represented by the Formula [VII] as defined in claim 14, which comprises reacting a naphthalene compound represented by the following Formula [IX] with an N-halogenated succinimide represented by the following Formula [X]:

[Chemical Formula 34]

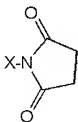
Formula [IX]



in the Formula [IX], where R⁷⁴ and R⁷⁵ may be the same or different, at least one of R⁷⁴ and R⁷⁵ represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom; R⁷⁶ represents a hydrogen atom, a substituted or unsubstituted, saturated or unsaturated alkyl group, a substituted or unsubstituted, alicyclic hydrocarbon group, a substituted or unsubstituted aryl group, a substituted or unsubstituted alkoxy group, a substituted or unsubstituted, alicyclic hydrocarbyloxy group, or a substituted or unsubstituted, aromatic hydrocarbyloxy group; and X represents a halogen atom;

[Chemical Formula 35]

Formula [X]



in the Formula [X], where X represents a halogen atom.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Pertains]

This invention relates to organic electroluminescent devices, especially organic electroluminescent devices containing aminostyrylnaphthalene compounds useful as electron transport materials, hole transport materials or light emitting materials, the aminostyrylnaphthalene compounds useful in the organic electroluminescent devices and their synthesis intermediates, and production processes of these compounds and intermediates.

[0002]

[Background Art]

In recent years, organic electroluminescent devices (EL devices) are attracting attention as a candidate for flat panel displays which can produce natural light, have a high response speed and have no visibility angle dependency, and accordingly, there is an increasing interest on organic materials as their constituents. Among such organic materials, however, there are not many materials capable of forming stable

red-light emitting layers in particular. Finding of such materials has, therefore, become an indispensable requirement for the realization of full-color, organic electroluminescent devices.

[0003]

As light-sensitive materials for electrophotography, aminostyryl compounds are disclosed, for example, in Japanese Patent Laid-open No.: Hei 5-105645, 2001-051433, 2002-131943, 2002-116560, 2002-099103, 2002-072511, 2002-040677, 2002-040676, 2002-031901, 2001-337469, 2001-337649, and 2000-214610. However, these compounds contain no electron attracting group in their molecules and hence, cannot be used for such applications as red-color emitting materials for organic electroluminescent devices.

[0004]

As materials for organic electroluminescent devices, certain compounds are disclosed in Japanese Patent Laid-open No. Hei 3-200889, Hei 5-194943, and 2002-226722. As illustrative materials employed in white-light organic electroluminescent devices, other materials are disclosed in Japanese Patent Laid-open No. Hei 6-207170. These materials are, however, not for the emission of red light either. Further, materials with

one or more styryl groups contained in combination with one or more triphenylamino groups are proposed in Japanese Patent Laid-open No.: Hei 5-320632, Hei 6-100857, Hei 9-268284, Hei 11-040359, Hei 11-102784, and Hei 10-245549. These materials, however, cannot be used for the emission of red light either.

[0005]

Aminostyryl compounds useful as red-light emitting materials in organic electroluminescent devices are led by the aminostyryl compounds disclosed in Inorganic and Organic Electroluminescence '96 Berlin, p.101, 1996; Journal of the Korean Chemical Society (1999), 43(3), 315-320; Bulletin of the Korean Chemical Society (2001), 22(2), 228-230; and Journal of the Korean Chemical Society (1999), 43(3), 315-320, and include those disclosed in Japanese Patent Laid-open No.: 2000-230132, 2002-022672, 2001-2883772, 2001-106657, and 2001-106658. Further, their application examples are reported in Japanese Patent Laid-open No.: Hei 11-329730, Hei 11-329731, 2000-012225, 2000-012228, 2000-012227, 2000-012226, 2001-305754, and 2000-136168. As described in Japanese Patent Laid-open No. 2002-134276, 2001-291591, 2001-307884 and 2001-307885, two or more of these materials may be positively combined together for use.

[0006]

As the molecular structures of the materials referred to in the above, many of them have structures symmetrical relative to the molecular long axes thereof. To obtain an emission maximum at an optimal wavelength or to permit the exhibition of improved evaporation upon fabrication of organic electroluminescent devices many of which are fabricated by vacuum evaporation, however, asymmetrical structures may be effective in certain instances as disclosed in Japanese Patent Laid-open No.: 2002-226722, 2001-288377, 2001-110570 (Patent Document 1), 2001-110571 and 2000-173773.

[0007]

Japanese Patent Laid-open No. 2002-208488, on the other hand, discloses that such asymmetrical structures are also effective as structural units for polymers. Further, their applications as multiphoton absorbers are also considered to be promising as disclosed in Science (1998), 281(11), 1653; WO 2001-096409; NATO Science Series, 3: High Technology (2000), 79 (Multiphoton and Light Driven Multielectron Processes in Organics), 53-65; Journal of Chemical Physics (2000), 113(10), 3951-3959; Journal of Physical Chemistry A (2001), 105(51), 11488-11495; Polymer Preprints (American Chemical Society,

Division of Polymer Chemistry) (1998), 39(2), 1116; and Materials Research Society Symposium Proceedings (1998), 488 (Electrical, Optical, and Magnetic Properties of Organic Solid-State Materials IV), 217-226.

[0008]

It is difficult to develop stable, high-luminance red-light emitting devices. Examples of those reported to date include a red-light emitting device making use of tris(8-quinolinato)aluminum (hereinafter abbreviated as "Alq₃") doped with 4-dicyanomethylene-6-(p-dimethylaminostyryl)-2-methyl-4H-pyran (hereinafter abbreviated as "DCM") (Chem. Funct. Dyes, Proc. Int. Symp., 2nd, p.536, 1993). As an example having achieved a reduction in the high crystallinity of DCM, there is 4-dicyanomethylene-6-(p-dimethylaminostyryl)-2-(t-butyl)-4H-pyran (hereinafter abbreviated as "DCJTb") disclosed in Macromol. Symp., 125, 49, 1997. However, their reliability such as service life is not satisfactory as display materials.

[0009]

[Patent Document 1]

Japanese Patent Laid-open No. 2001-110570 (page 4, right column, line 40 to page 5, right column, line 4 from the bottom; page 7, right column, line 30 to page 8,

left column, line 17; FIG. 1 to FIG. 8)

[0010]

[Problems to be Solved by the Invention]

In the development of organic electroluminescent devices, selection of light emitting materials is the most important theme in assuring reliability for the devices. The aminostyrylnaphthalene compounds disclosed in Patent Document 1 are excellent in color purity and high in fluorescence quantum yield and moreover, can form stable, amorphous thin films. Nonetheless, it is the current situation that there is still an outstanding desire for the realization of a red-light emitting device of high luminance, high stability and high color purity.

[0011]

Objects of the present invention are to provide an organic electroluminescent device capable of producing stable and high-luminance emission of red light at an optimal wavelength by using a compound improved in fluorescent wavelength over an aminostyrylnaphthalene compound which can produce emission of red light at a high fluorescence quantum yield, an aminostyrylnaphthalene compound useful in the organic electroluminescent device and its synthesis intermediates, and production processes of these compound and

intermediates.

[0012]

[Means for Solving the Problems]

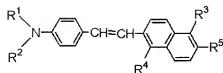
The present inventors have proceeded with an extensive investigation to achieve the above-described objects. As a result, it has been found that fabrication of an organic electroluminescent device with a luminescent layer, which makes use of a particular styryl compound in combination with a material capable of efficiently transmitting energy especially to the styryl compound, can provide a red-light emitting device of still higher luminance and reliability, leading to the present invention.

[0013]

Described specifically, the present invention relates to an organic electroluminescent device including an organic layer having a light-emitting area and arranged between an anode and a cathode, wherein at least a part of the organic layer includes at least one aminostyrylnaphthalene compound represented by the below-described Formula [I], [II] and [III]; and also to the aminostyrylnaphthalene compound:

[Chemical Formula 36]

Formula [I]



in the Formula [I], where

R^1 and R^2 may be the same or different and each independently represents a phenyl group represented by the following Formula (1),

[Chemical Formula 37]

Formula (1)



in the Formula (1), where R^6 , R^7 , R^8 , R^9 and R^{10} may be the same or different, at least one of R^6 to R^{10} represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom with a proviso that, the two adjacent ones of R^6 to R^{10} may be fused together to form a ring, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^6 to R^{10} are each a hydrogen atom,

R^3 and R^4 may be the same or different, at least

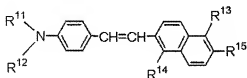
one of R³ and R⁴ represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R⁵ represents a substituted or unsubstituted, saturated or unsaturated alkyl group such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl or allyl, a substituted or unsubstituted, alicyclic hydrocarbon group such as cyclohexyl, a substituted or unsubstituted aryl group such as phenyl, naphthyl or anthranyl, a substituted or unsubstituted alkoxyl group such as methoxy, ethoxy, n-propoxy, i-propoxy, n-butoxy, i-butoxy or t-butoxy, a substituted or unsubstituted, alicyclic hydrocarbyloxy group such as cyclohexyloxy, or a substituted or unsubstituted, aromatic hydrocarbyloxy group such as phenoxy, naphthoxy or anthroxy.

[0014]

[Chemical Formula 38]

Formula [II]



in the Formula [II], where

R¹¹ and R¹² may be the same or different and each

independently represents a naphthyl group represented by the following Formula (2),

[Chemical Formula 39]

Formula (2)



or

in the Formula (2), where R¹⁶, R¹⁷, R¹⁸, R¹⁹, R²⁰, R²¹ and R²² may be the same or different, at least one of R¹⁶ to R²² represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R¹⁶ to R²² are each a hydrogen atom,

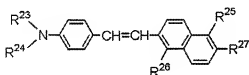
R¹³ and R¹⁴ may be the same or different, at least one of R¹³ and R¹⁴ represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R¹⁵ represents a hydrogen atom, a substituted or unsubstituted, saturated or unsaturated alkyl group such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl or allyl, a substituted or unsubstituted, alicyclic hydrocarbon group such as cyclohexyl, a substituted or unsubstituted aryl group such as phenyl, naphthyl or anthranyl, a substituted or unsubstituted alkoxy group such as methoxy, ethoxy, n-propoxy, i-propoxy, n-butoxy, i-butoxy or t-butoxy, a substituted or unsubstituted, alicyclic hydrocarbyloxy group such as cyclohexyloxy, or a substituted or unsubstituted, aromatic hydrocarbyloxy group such as phenoxy, naphthoxy or anthroxy.

[0015]

[Chemical Formula 40]

Formula [III]



in the Formula [III], where

R²³ is a phenyl group represented by the following Formula (3),

[Chemical Formula 41]

Formula (3)

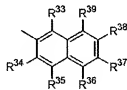
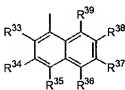


in the Formula (3), where R²⁸, R²⁹, R³⁰, R³¹ and R³² may be the same or different, at least one of R²⁸ to R³¹ represents a hydrogen atom, a saturated or unsaturated hydrocarbon group having at least one carbon atom with a proviso that, the two adjacent ones of R²⁶ to R³¹ may be fused together to form a ring, a hydrocarbyloxy group having at least one carbon atom, a hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R²⁸ to R³¹ are each a hydrogen atom,

R²⁴ represents a naphthyl group represented by the following Formula (4),

[Chemical Formula 42]

Formula (4)



or

in the Formula (4), where R³³, R³⁴, R³⁵, R³⁶, R³⁷, R³⁸ and R³⁹ may be the same or different, at least one of R³³ to R³⁹ represents a hydrogen atom, a saturated or

unsaturated hydrocarbon group having at least one carbon atom, a saturated or unsaturated hydrocarbyloxy group having at least one carbon atom, a saturated or unsaturated hydrocarbylamino group having at least one carbon atom, a trifluoromethyl group, a cyano group or a halogen atom, and the remaining one or ones of R^{33} to R^{39} are each a hydrogen atom,

R^{25} and R^{26} may be the same or different, at least one of R^{25} and R^{26} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and

R^{27} represents a hydrogen atom, a substituted or unsubstituted, saturated or unsaturated alkyl group such as methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl or allyl, a substituted or unsubstituted, alicyclic hydrocarbon group such as cyclohexyl, a substituted or unsubstituted aryl group such as phenyl, naphthyl or anthranyl, a substituted or unsubstituted alkoxy group such as methoxy, ethoxy, n-propoxy, i-propoxy, n-butoxy, i-butoxy or t-butoxy, a substituted or unsubstituted, alicyclic hydrocarbyloxy group such as cyclohexyloxy, or a substituted or unsubstituted,

aromatic hydrocarbyloxy group such as phenoxy, naphthoxy or anthroxy.

[0016]

According to the organic electroluminescent device of the present invention, the aminostyrylnaphthalene compound has the specific structure represented by the formula [I], [II] or [III]. Owing to its specific structure, the compound is excellent especially in the emission of red light, and is equipped with electron transporting ability based on one or more electron attracting groups such as one or more cyano groups on a naphthalene group and also with hole transporting ability based on an aminostyryl group, and moreover, shows amorphous properties advantageous for film formability by vacuum deposition or the like and also durability. Use of the aminostyrylnaphthalene compound, therefore, can provide an organic electroluminescent device capable of producing high-luminance and stable emission of red light at an optimal wavelength.

[0017]

The organic electroluminescent device according to the present invention preferably has the organic layer is in a form of an organic layer structure including a

hole transport layer and an electron transport layer stacked one over the other, and at least the electron transport layer in the organic layer includes at least the one aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III].

[0018]

In addition, preferably, the organic layer is in a form of an organic layer structure including a hole transport layer and at least the hole transport layer in the organic layer includes at least the one aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III].

[0019]

In addition, preferably, the organic layer is in a form of an organic layer structure including a hole transport layer, a luminescent layer and an electron transport layer stacked one over another, and at least the luminescent layer in the organic layer includes at least the one aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III].

[0020]

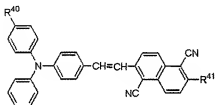
In addition, preferably, the aminostyrylnaphthalene compound is represented by the following Formula (5), (6), (7), (8), (9), (10), (11),

(12), (13), (14), (15), (16) or (17).

[0021]

[Chemical Formula 43]

Formula (5)

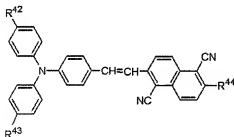


in the Formula (5), where R⁴⁰ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁴¹ has the same meaning as R⁵.

[0022]

[Chemical Formula 44]

Formula (6)



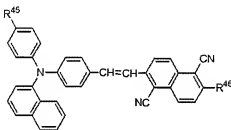
in the Formula (6), where R⁴² and R⁴³ may be the same or different and each independently represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and

R⁴⁴ has the same meaning as R⁵.

[0023]

[Chemical Formula 45]

Formula (7)

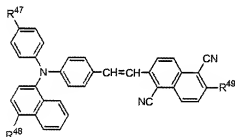


in the Formula (7), where R⁴⁵ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁴⁶ has the same meaning as R²⁷.

[0024]

[Chemical Formula 46]

Formula (8)



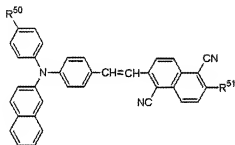
in the Formula (8), where R⁴⁷ and R⁴⁸ may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁴⁹ has the same meaning as

R²⁷.

[0025]

[Chemical Formula 47]

Formula (9)

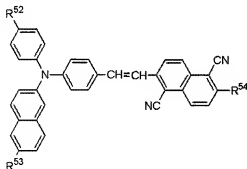


in the Formula (9), where R⁵⁰ represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R⁵¹ has the same meaning as R²⁷.

[0026]

[Chemical Formula 48]

Formula (10)



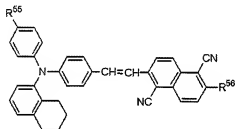
in the Formula (10), where R⁵² and R⁵³ may be the same or different, a saturated or unsaturated alkyl group

having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R^{54} has the same meaning as R^{27} .

[0027]

[Chemical Formula 49]

Formula (11)

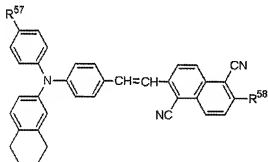


in the Formula (11), where R^{55} represents a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R^{56} has the same meaning as R^5 .

[0028]

[Chemical Formula 50]

Formula (12)



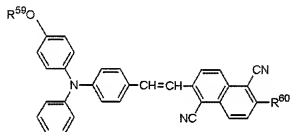
in the Formula (12), where R^{57} represents a saturated or unsaturated alkyl group having 1 to 6 carbon

atoms or a substituted or unsubstituted aryl group, and
 R^{58} has the same meaning as R^5 .

[0029]

[Chemical Formula 51]

Formula (13)

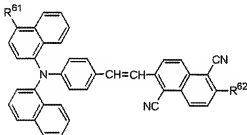


in the Formula (13), where R^{59} represents a
saturated or unsaturated alkyl group having 1 to 6 carbon
atoms or a substituted or unsubstituted aryl group, and
 R^{60} has the same meaning as R^5 .

[0030]

[Chemical Formula 52]

Formula (14)



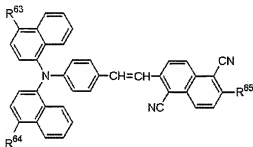
in the Formula (14), where R^{61} represents a
saturated or unsaturated alkyl group having 1 to 6 carbon

atoms or a substituted or unsubstituted aryl group, and R^{62} has the same meaning as R^{15} .

[0031]

[Chemical Formula 53]

Formula (15)

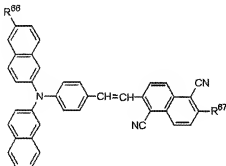


in the Formula (15), where R^{63} and R^{64} may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R^{65} has the same meaning as R^{15} .

[0032]

[Chemical Formula 54]

Formula (16)



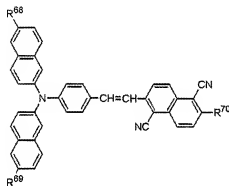
in the Formula (16), where R^{66} represents a

saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R^{67} has the same meaning as R^{15} .

[0033]

[Chemical Formula 55]

Formula (17)



in the Formula (17), where R^{68} and R^{69} may be the same or different, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms or a substituted or unsubstituted aryl group, and R^{70} has the same meaning as R^{15} .

[0034]

The present invention also provides an aminostyrylnaphthalene compound represented by the above-described formula [I], [II] or [III].

[0035]

The above-described compound of the present invention can be effectively used as an organic light-

emitting material capable of producing emission of red light of good chromaticity at a relatively-short, fluorescent wavelength. Further, it has a relatively small molecular weight and therefore, can reduce a thermal load to be applied upon vacuum evaporation or the like. Moreover, it is superb in electrical, thermal or chemical stability, and is amorphous and can readily form a glass state. Accordingly, it permits vacuum deposition or the like. With the organic electroluminescent device making use of the compound according to the present invention, red light the wavelength of which is relatively short is emitted. Therefore, the organic electroluminescent device is also advantageous in obtaining resonant light of improved color purity when the resonator structure disclosed in International publication No. WO 01/39554 is fabricated.

[0036]

The compound according to the present invention may preferably be represented by any one of the above-described formulas (5) to (17).

[0037]

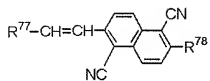
Representing the preferred compound by the following formula [I'], the preferred compound can be exemplified as will be presented below in Table 1 to

Table 24.

[0038]

[Chemical Formula 56]









Formula [I']



wherein R⁷⁷ and R⁷⁸ are as will be specified in the following tables which show diverse combinations of R⁷⁷ and R⁷⁸.








[0039]

[Table 1]

	R^{78}							
	-H	-CH ₃	-C ₂ H ₅	<i>n</i> -C ₃ H ₇	<i>i</i> -C ₃ H ₇	<i>n</i> -C ₄ H ₉	<i>i</i> -C ₄ H ₉	<i>t</i> -C ₄ H ₉
	(20)-1	(20)-19	(20)-37	(20)-55	(20)-73	(20)-91	(20)-109	(20)-127
	(20)-2	(20)-20	(20)-38	(20)-56	(20)-74	(20)-92	(20)-110	(20)-128
	(20)-3	(20)-21	(20)-39	(20)-57	(20)-75	(20)-93	(20)-111	(20)-129
	(20)-4	(20)-22	(20)-40	(20)-58	(20)-76	(20)-94	(20)-112	(20)-130
	(20)-5	(20)-23	(20)-41	(20)-59	(20)-77	(20)-95	(20)-113	(20)-131
	(20)-6	(20)-24	(20)-42	(20)-60	(20)-78	(20)-96	(20)-114	(20)-132
	(20)-7	(20)-25	(20)-43	(20)-61	(20)-79	(20)-97	(20)-115	(20)-133
	(20)-8	(20)-26	(20)-44	(20)-62	(20)-80	(20)-98	(20)-116	(20)-134




[0040]

[Table 2]

	R^{70}							
	-H	-CH ₃	-C ₂ H ₅	- <i>n</i> -C ₃ H ₇	- <i>i</i> -C ₃ H ₇	- <i>n</i> -C ₄ H ₉	- <i>i</i> -C ₄ H ₉	- <i>t</i> -C ₄ H ₉
	(20)-9	(20)-27	(20)-45	(20)-63	(20)-81	(20)-99	(20)-117	(20)-135
	(20)-10	(20)-28	(20)-46	(20)-64	(20)-82	(20)-100	(20)-118	(20)-136
	(20)-11	(20)-29	(20)-47	(20)-65	(20)-83	(20)-101	(20)-119	(20)-137
	(20)-12	(20)-30	(20)-48	(20)-66	(20)-84	(20)-102	(20)-120	(20)-138
	(20)-13	(20)-31	(20)-49	(20)-67	(20)-85	(20)-103	(20)-121	(20)-139
	(20)-14	(20)-32	(20)-50	(20)-68	(20)-86	(20)-104	(20)-122	(20)-140
	(20)-15	(20)-33	(20)-51	(20)-69	(20)-87	(20)-105	(20)-123	(20)-141
















[0041]

[Table 3]

	R^{78}							
	-H	-CH ₃	-C ₂ H ₅	<i>m</i> -C ₃ H ₇	<i>p</i> -C ₃ H ₇	<i>m</i> -C ₄ H ₉	<i>p</i> -C ₄ H ₉	<i>t</i> -C ₄ H ₉
	(20)-16	(20)-34	(20)-52	(20)-70	(20)-88	(20)-106	(20)-124	(20)-142
	(20)-17	(20)-35	(20)-53	(20)-71	(20)-89	(20)-107	(20)-125	(20)-143
	(20)-18	(20)-36	(20)-54	(20)-72	(20)-90	(20)-108	(20)-126	(20)-144
















[0042]

[Table 4]

		R ^{7B}							
									
R ⁷⁷		(21)-1	(21)-19	(21)-37	(21)-55	(21)-73	(21)-91	(21)-109	(21)-127
		(21)-2	(21)-20	(21)-38	(21)-56	(21)-74	(21)-92	(21)-110	(21)-128
		(21)-3	(21)-21	(21)-39	(21)-57	(21)-75	(21)-93	(21)-111	(21)-129
		(21)-4	(21)-22	(21)-40	(21)-58	(21)-76	(21)-94	(21)-112	(21)-130
		(21)-5	(21)-23	(21)-41	(21)-59	(21)-77	(21)-95	(21)-113	(21)-131
		(21)-6	(21)-24	(21)-42	(21)-60	(21)-78	(21)-96	(21)-114	(21)-132
		(21)-7	(21)-25	(21)-43	(21)-61	(21)-79	(21)-97	(21)-115	(21)-133

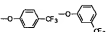










[0043]

[Table 5]

		R ^{7B}							
									
R ⁷⁷		(21)-8	(21)-26	(21)-44	(21)-62	(21)-80	(21)-98	(21)-116	(21)-134
		(21)-9	(21)-27	(21)-45	(21)-63	(21)-81	(21)-99	(21)-117	(21)-135
		(21)-10	(21)-28	(21)-46	(21)-64	(21)-82	(21)-100	(21)-118	(21)-136
		(21)-11	(21)-29	(21)-47	(21)-65	(21)-83	(21)-101	(21)-119	(21)-137
		(21)-12	(21)-30	(21)-48	(21)-66	(21)-84	(21)-102	(21)-120	(21)-138
		(21)-13	(21)-31	(21)-49	(21)-67	(21)-85	(21)-103	(21)-121	(21)-139
		(21)-14	(21)-32	(21)-50	(21)-68	(21)-86	(21)-104	(21)-122	(21)-140

[0044]

[Table 6]

		R ^{7B}							
									
R ⁷⁷		(21)-15	(21)-33	(21)-51	(21)-69	(21)-87	(21)-105	(21)-123	(21)-141
		(21)-16	(21)-34	(21)-52	(21)-70	(21)-88	(21)-106	(21)-124	(21)-142
		(21)-17	(21)-35	(21)-53	(21)-71	(21)-89	(21)-107	(21)-125	(21)-143
		(21)-18	(21)-36	(21)-54	(21)-72	(21)-90	(21)-108	(21)-126	(21)-144
















[0045]

[Table 7]

		R ⁷⁸							
R ⁷⁷		(22)-1	(22)-19	(22)-37	(22)-55	(22)-73	(22)-91	(22)-109	(22)-127
		(22)-2	(22)-20	(22)-38	(22)-56	(22)-74	(22)-92	(22)-110	(22)-128
		(22)-3	(22)-21	(22)-39	(22)-57	(22)-75	(22)-93	(22)-111	(22)-129
		(22)-4	(22)-22	(22)-40	(22)-58	(22)-76	(22)-94	(22)-112	(22)-130
		(22)-5	(22)-23	(22)-41	(22)-59	(22)-77	(22)-95	(22)-113	(22)-131
		(22)-6	(22)-24	(22)-42	(22)-60	(22)-78	(22)-96	(22)-114	(22)-132
		(22)-7	(22)-25	(22)-43	(22)-61	(22)-79	(22)-97	(22)-115	(22)-133













[0046]

[Table 8]

		R ⁷⁸							
									
R ⁷⁷		(22)-8	(22)-26	(22)-44	(22)-62	(22)-80	(22)-98	(22)-116	(22)-134
		(22)-9	(22)-27	(22)-45	(22)-63	(22)-81	(22)-99	(22)-117	(22)-135
		(22)-10	(22)-28	(22)-46	(22)-64	(22)-82	(22)-100	(22)-118	(22)-136
		(22)-11	(22)-29	(22)-47	(22)-65	(22)-83	(22)-101	(22)-119	(22)-137
		(22)-12	(22)-30	(22)-48	(22)-66	(22)-84	(22)-102	(22)-120	(22)-138
		(22)-13	(22)-31	(22)-49	(22)-67	(22)-85	(22)-103	(22)-121	(22)-139
		(22)-14	(22)-32	(22)-50	(22)-68	(22)-86	(22)-104	(22)-122	(22)-140

[0047]

[Table 9]

	R^{78}							
								
R^{77} 	(22)-15	(22)-33	(22)-51	(22)-69	(22)-87	(22)-105	(22)-123	(22)-141
	(22)-16	(22)-34	(22)-52	(22)-70	(22)-88	(22)-106	(22)-124	(22)-142
	(22)-17	(22)-35	(22)-53	(22)-71	(22)-89	(22)-107	(22)-125	(22)-143
	(22)-18	(22)-36	(22)-54	(22)-72	(22)-90	(22)-108	(22)-126	(22)-144












[0048]

[Table 10]

		R ⁷⁸							
						-OCH ₃	-OC ₂ H ₅	-O(<i>n</i> -C ₃ H ₇)	-O(<i>i</i> -C ₃ H ₇)
R ⁷⁷		(23)-1	(23)-19	(23)-37	(23)-55	(23)-73	(23)-91	(23)-109	(23)-127
		(23)-2	(23)-20	(23)-38	(23)-56	(23)-74	(23)-92	(23)-110	(23)-128
		(23)-3	(23)-21	(23)-39	(23)-57	(23)-75	(23)-93	(23)-111	(23)-129
		(23)-4	(23)-22	(23)-40	(23)-58	(23)-76	(23)-94	(23)-112	(23)-130
		(23)-5	(23)-23	(23)-41	(23)-59	(23)-77	(23)-95	(23)-113	(23)-131
		(23)-6	(23)-24	(23)-42	(23)-60	(23)-78	(23)-96	(23)-114	(23)-132
		(23)-7	(23)-25	(23)-43	(23)-61	(23)-79	(23)-97	(23)-115	(23)-133









[0049]

[Table 11]

		R ^{7B}							
						-OCH ₃	-OC ₂ H ₅	-O(<i>n</i> -C ₃ H ₇)	-O(<i>i</i> -C ₃ H ₇)
R ⁷⁷		(23)-8	(23)-26	(23)-44	(23)-62	(23)-80	(23)-98	(23)-116	(23)-134
		(23)-9	(23)-27	(23)-45	(23)-63	(23)-81	(23)-99	(23)-117	(23)-135
		(23)-10	(23)-28	(23)-46	(23)-64	(23)-82	(23)-100	(23)-118	(23)-136
		(23)-11	(23)-29	(23)-47	(23)-65	(23)-83	(23)-101	(23)-119	(23)-137
		(23)-12	(23)-30	(23)-48	(23)-66	(23)-84	(23)-102	(23)-120	(23)-138
		(23)-13	(23)-31	(23)-49	(23)-67	(23)-85	(23)-103	(23)-121	(23)-139
		(23)-14	(23)-32	(23)-50	(23)-68	(23)-86	(23)-104	(23)-122	(23)-140












[0050]

[Table 12]

R ⁷⁷	R ⁷⁸							
					-OCH ₃	-OC ₂ H ₅	-O(<i>n</i> -C ₃ H ₇)	-O(<i>i</i> -C ₃ H ₇)
	(23)-15	(23)-33	(23)-51	(23)-69	(23)-87	(23)-105	(23)-123	(23)-141
	(23)-16	(23)-34	(23)-52	(23)-70	(23)-68	(23)-106	(23)-124	(23)-142
	(23)-17	(23)-35	(23)-53	(23)-71	(23)-89	(23)-107	(23)-125	(23)-143
	(23)-18	(23)-36	(23)-54	(23)-72	(23)-90	(23)-108	(23)-126	(23)-144













[0051]

[Table 13]

	R ⁷⁸								
	-O(<i>n</i> -C ₄ H ₉)	-O(<i>i</i> -C ₄ H ₉)	-O(<i>t</i> -C ₄ H ₉)	-O- 	-O- 	-O- 	-O- 	-O-	
R ⁷⁷		(24)-1	(24)-19	(24)-37	(24)-55	(24)-73	(24)-91	(24)-109	(24)-127
		(24)-2	(24)-20	(24)-38	(24)-56	(24)-74	(24)-92	(24)-110	(24)-128
		(24)-3	(24)-21	(24)-39	(24)-57	(24)-75	(24)-93	(24)-111	(24)-129
		(24)-4	(24)-22	(24)-40	(24)-58	(24)-76	(24)-94	(24)-112	(24)-130
		(24)-5	(24)-23	(24)-41	(24)-59	(24)-77	(24)-95	(24)-113	(24)-131
		(24)-6	(24)-24	(24)-42	(24)-60	(24)-78	(24)-96	(24)-114	(24)-132
		(24)-7	(24)-25	(24)-43	(24)-61	(24)-79	(24)-97	(24)-115	(24)-133










[0052]

[Table 14]

	R ⁷⁸							
	-O(<i>n</i> -C ₄ H ₉)	-O(<i>i</i> -C ₄ H ₉)	-O(<i>t</i> -C ₄ H ₉)	-O-  -O-  -CH ₃	-O-  -CH ₃	-O-  -C ₂ H ₅	-O-  - <i>n</i> -C ₃ H ₇	
	(24)-8	(24)-26	(24)-44	(24)-62	(24)-80	(24)-98	(24)-116	(24)-134
	(24)-9	(24)-27	(24)-45	(24)-63	(24)-81	(24)-99	(24)-117	(24)-135
	(24)-10	(24)-28	(24)-46	(24)-64	(24)-82	(24)-100	(24)-118	(24)-136
	(24)-11	(24)-29	(24)-47	(24)-65	(24)-83	(24)-101	(24)-119	(24)-137
	(24)-12	(24)-30	(24)-48	(24)-66	(24)-84	(24)-102	(24)-120	(24)-138
	(24)-13	(24)-31	(24)-49	(24)-67	(24)-85	(24)-103	(24)-121	(24)-139
	(24)-14	(24)-32	(24)-50	(24)-68	(24)-86	(24)-104	(24)-122	(24)-140














[0053]

[Table 15]

R ⁷⁷	R ⁷⁸							
	-O(<i>n</i> -C ₄ H ₉)	-O(<i>i</i> -C ₄ H ₉)	-O(<i>t</i> -C ₄ H ₉)	-O- 	-O-  -CH ₃	-O- 	-O- 	-O- 
	(24)-15	(24)-33	(24)-51	(24)-69	(24)-87	(24)-105	(24)-123	(24)-141
	(24)-16	(24)-34	(24)-52	(24)-70	(24)-88	(24)-106	(24)-124	(24)-142
	(24)-17	(24)-35	(24)-53	(24)-71	(24)-89	(24)-107	(24)-125	(24)-143
	(24)-18	(24)-36	(24)-54	(24)-72	(24)-90	(24)-108	(24)-126	(24)-144















[0054]

[Table 16]

	R ⁷⁸								
									
R ⁷⁷		(25)-1	(25)-19	(25)-37	(25)-55	(25)-73	(25)-91	(25)-109	(25)-127
		(25)-2	(25)-20	(25)-38	(25)-56	(25)-74	(25)-92	(25)-110	(25)-128
		(25)-3	(25)-21	(25)-39	(25)-57	(25)-75	(25)-93	(25)-111	(25)-129
		(25)-4	(25)-22	(25)-40	(25)-58	(25)-76	(25)-94	(25)-112	(25)-130
		(25)-5	(25)-23	(25)-41	(25)-59	(25)-77	(25)-95	(25)-113	(25)-131
		(25)-6	(25)-24	(25)-42	(25)-60	(25)-78	(25)-96	(25)-114	(25)-132
		(25)-7	(25)-25	(25)-43	(25)-61	(25)-79	(25)-97	(25)-115	(25)-133

[0055]

[Table 17]

		R ^{7b}							
									
R ^{7a}		(25)-8	(25)-26	(25)-44	(25)-62	(25)-80	(25)-98	(25)-116	(25)-134
		(25)-9	(25)-27	(25)-45	(25)-63	(25)-81	(25)-99	(25)-117	(25)-135
		(25)-10	(25)-28	(25)-46	(25)-64	(25)-82	(25)-100	(25)-118	(25)-136
		(25)-11	(25)-29	(25)-47	(25)-65	(25)-83	(25)-101	(25)-119	(25)-137
		(25)-12	(25)-30	(25)-48	(25)-66	(25)-84	(25)-102	(25)-120	(25)-138
		(25)-13	(25)-31	(25)-49	(25)-67	(25)-85	(25)-103	(25)-121	(25)-139
		(25)-14	(25)-32	(25)-50	(25)-68	(25)-86	(25)-104	(25)-122	(25)-140

[0056]

[Table 18]

	R^{7B}								
R^{7A}		(25)-15	(25)-33	(25)-51	(25)-69	(25)-87	(25)-105	(25)-123	(25)-141
		(25)-16	(25)-34	(25)-52	(25)-70	(25)-88	(25)-106	(25)-124	(25)-142
		(25)-17	(25)-35	(25)-53	(25)-71	(25)-89	(25)-107	(25)-125	(25)-143
		(25)-18	(25)-36	(25)-54	(25)-72	(25)-90	(25)-108	(25)-126	(25)-144
















[0057]

[Table 19]

R^{77}	R^{78}							
	(26)-1	(26)-19	(26)-37	(26)-55	(26)-73	(26)-91	(26)-109	(26)-127
	(26)-2	(26)-20	(26)-38	(26)-56	(26)-74	(26)-92	(26)-110	(26)-128
	(26)-3	(26)-21	(26)-39	(26)-57	(26)-75	(26)-93	(26)-111	(26)-129
	(26)-4	(26)-22	(26)-40	(26)-58	(26)-76	(26)-94	(26)-112	(26)-130
	(26)-5	(26)-23	(26)-41	(26)-59	(26)-77	(26)-95	(26)-113	(26)-131
	(26)-6	(26)-24	(26)-42	(26)-60	(26)-78	(26)-96	(26)-114	(26)-132
	(26)-7	(26)-25	(26)-43	(26)-61	(26)-79	(26)-97	(26)-115	(26)-133













[0058]

[Table 20]

		R ^{7B}							
									
R ⁷⁷		(26)-8	(26)-26	(26)-44	(26)-62	(26)-80	(26)-98	(26)-116	(26)-134
		(26)-9	(26)-27	(26)-45	(26)-63	(26)-81	(26)-99	(26)-117	(26)-135
		(26)-10	(26)-28	(26)-46	(26)-64	(26)-82	(26)-100	(26)-118	(26)-136
		(26)-11	(26)-29	(26)-47	(26)-65	(26)-83	(26)-101	(26)-119	(26)-137
		(26)-12	(26)-30	(26)-48	(26)-66	(26)-84	(26)-102	(26)-120	(26)-138
		(26)-13	(26)-31	(26)-49	(26)-67	(26)-85	(26)-103	(26)-121	(26)-139
		(26)-14	(26)-32	(26)-50	(26)-68	(26)-86	(26)-104	(26)-122	(26)-140










[0059]

[Table 21]

R ⁷⁷	R ⁷⁸							
								
	(26)-15	(26)-33	(26)-51	(26)-69	(26)-87	(26)-105	(26)-123	(26)-141
	(26)-16	(26)-34	(26)-52	(26)-70	(26)-88	(26)-106	(26)-124	(26)-142
	(26)-17	(26)-35	(26)-53	(26)-71	(26)-89	(26)-107	(26)-125	(26)-143
	(26)-18	(26)-36	(26)-54	(26)-72	(26)-90	(26)-108	(26)-126	(26)-144










[0060]

[Table 22]

	R ⁷⁸	
		
R ⁷⁷		(27)-1 (27)-19
		(27)-2 (27)-20
		(27)-3 (27)-21
		(27)-4 (27)-22
		(27)-5 (27)-23
		(27)-6 (27)-24
		(27)-7 (27)-25







[0061]

[Table 23]

	R ⁷⁸	
		
R ⁷⁷		(27)-8 (27)-26
		(27)-9 (27)-27
		(27)-10 (27)-28
		(27)-11 (27)-29
		(27)-12 (27)-30
		(27)-13 (27)-31
		(27)-14 (27)-32

[0062]

[Table 24]

	R^{78}	
		
R^{77}		
	(27)-15	(27)-33
	(27)-16	(27)-34
	(27)-17	(27)-35
	(27)-18	(27)-36

[0063]

Examples of materials usable for the formation of the organic electroluminescent device according to the present invention can include, in addition to the compounds according to the present invention, hole transport materials (for example, aromatic amines and the like), electron transport materials (for example, Alq_3 , pyrazoline and the like), or a series of compounds commonly employed as red emission dopants (DCM and its analogous compounds, porphyrins, phthalocyanines, perylene compounds, Nile Red, squalilium compounds, and

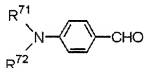
the like).

[0064]

As a process for producing the compounds of the present invention with high efficiency, the present invention also provides a process for the production of an aminostyrylnaphthalene compound represented by the Formula [I], [II] or [III], which includes subjecting an aminobenzaldehyde represented by the following Formula [IV] and a phosphonate ester represented by the following Formula [V] and/or a phosphonium represented by the following Formula [VI] to condensation:

[Chemical Formula 57] ..

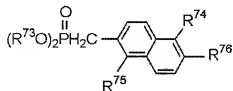
Formula [IV]



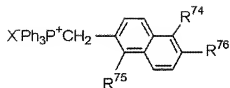
in the Formula [IV], where R⁷¹ and R⁷² each independently represents an aryl group corresponding to R¹, R², R¹¹, R¹², R²³ or R²⁴.

[Chemical Formula 58]

Formula [V]



Formula [VI]



in the Formulas [V] and [VI], where R^{73} represents a hydrocarbon group, preferably a saturated hydrocarbon group having 1 to 4 carbon atoms, R^{74} and R^{75} each independently represents a group corresponding to R^3 , R^4 , R^{13} , R^{14} , R^{25} or R^{26} , R^{76} represents a group corresponding to R^5 , R^{15} or R^{27} , and X represents a halogen atom such as F, Cl, Br and so on (hereinafter, ditto).

[0065]

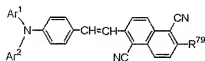
In a specific embodiment of the process according to the present invention for the production of the compound, the condensation may be conducted by the Wittig-Horner reaction or the Wittig reaction, at least one of the phosphonate ester and/or the phosphonium may be treated with a base in a solvent to form carbanions, and the carbanions and the 4-(N,N-diarylamino)benzaldehyde may be subjected to condensation.

[0066]

Upon obtaining, for example, an aminostyrylnaphthalene compound represented by the following formula (28):

[Chemical Formula 59]

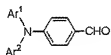
Formula (28)



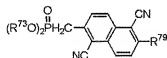
in the Formula (28), where Ar^1 and Ar^2 are each the same as R^1 , R^2 , R^{11} , R^{12} , R^{23} or R^{24} as defined above, and R^{79} is the same as R^5 , R^{15} or R^{27} as defined above, a 4-(N,N-diarylamino)benzaldehyde represented by the following Formula (29) and a phosphonate ester represented by the following formula (30) and/or a phosphonium represented by the following formula (31) are subjected to condensation.

[Chemical Formula 60]

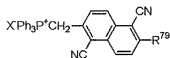
Formula (29)



Formula (30)



Formula (31)



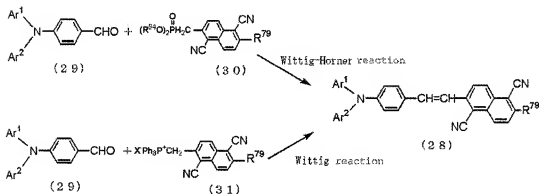
in the Formulas (29), (30) and (31), where Ar^1 , Ar^2 , R^{79} and X have the same meanings as defined above.

[0067]

Expressing the above reactions by a scheme, they can be shown, for example, as illustrated by the following reaction scheme 1:

[Chemical Formula 61]

Reaction Scheme 1



[0068]

The reactions firstly begin with the treatment of one of the compound of the Formula (30) and the compound of the Formula (31) with a base in an adequate solvent to produce carbanions. These carbanions and the aldehyde of the formula (29) are then subjected to condensation such that the reactions are completed. As examples of the combination of the base and the solvent, the following combinations can be mentioned.

[0069]

Sodium hydroxide/water, sodium carbonate/water, potassium carbonate/water, sodium ethoxide/ethanol or dimethylformamide, sodium methoxide/methanol-diethyl ether mixed solvent or dimethylformamide, triethylamine/ethanol or diglyme or chloroform or nitromethane, pyridine/methylene chloride or nitromethane, 1,5-disazabicyclo[4.3.0]non-5-ene/dimethylsulfoxide, potassium t-butoxide/dimethylsulfoxide or tetrahydrofuran or benzene or dimethylformamide, phenyllithium/diethyl ether or tetrahydrofuran, t-butyllithium/diethyl ether or tetrahydrofuran, sodium amide/ammonia, sodium hydride/dimethylformamide or tetrahydrofuran, triethylsodium/diethyl ether or tetrahydrofuran, and so on.

[0070]

As the reactions proceed at relatively low temperatures (-30°C to 30°C) and are selective, the target product can be readily purified by chromatography, and owing to the high crystallinity of the compound of the present invention represented by the Formula (28), its purity can be increased further by recrystallization. No particular limitation is imposed on the method of recrystallization, but the recrystallization may be

conveniently conducted by dissolving the compound in acetone and then adding hexane to the resulting solution or by dissolving the compound in toluene under heat, concentrating the thus-prepared solution and then cooling the resultant concentrate. The reactions may be brought to completion in 3 to 24 hours under normal pressure.

[0071]

By the production process according to the present invention, the aminostyrylnaphthalene compounds represented by the formulas (5) to (17), specifically the aminostyrylnaphthalene compounds represented by the formulas (20)-1 to (27)-36 can be obtained.

[0072]

The present invention also provides various compounds suitable as synthesis intermediates for the compound according to the present invention.

[0073]

Specifically, they include the phosphonate ester represented by the formula [V] or the phosphonium represented by the formula [VI], which is useful as a synthesis intermediate for the aminostyrylnaphthalene compound represented by the formula [I], [II] or [III].

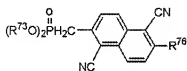
[0074]

The synthesis intermediate (hereinafter called

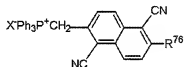
"the synthesis intermediate 1 of the present invention")
 is specifically represented by the following Formula (18)
 or (19):

[Chemical Formula 62]

Formula (18)



Formula (19)



in the Formulas (18), (19), where R^{73} , R^{76} and X
 have the same meanings as defined above.

[0075]

The synthesis intermediate 1 of the present
 invention can be derived from a synthesis intermediate 2
 as a precursor as will be described next.

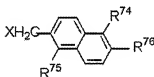
[0076]

The phosphonate ester represented by the Formula
 [V] or the phosphonium represented by the Formula [VI]
 can be obtained as a synthesis intermediate by reacting a
 halogenated aryl compound represented by the below-
 described Formula [VII] with a trialkyl phosphite

represented by the below-described Formula [VIII] or triphenylphosphine (PPh_3). This reaction can be brought to completion in 30 minutes to 24 hours at 120°C to 160°C under normal pressure in a solventless manner, in a solvent having a boiling point of 120°C or higher such as xylene, or in a large excess of a trialkyl phosphite:

[Chemical Formula 63]

Formula [VII]



in the Formula [VII], where R^{74} and R^{75} may be the same or different, at least one of R^{74} and R^{75} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and the remaining one represents a cyano group, a nitro group, a trifluoromethyl group or a halogen atom; R^{76} represents a hydrogen atom, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms, or a substituted or unsubstituted aryl group; and X represents a halogen atom;

Formula [VIII]



in the Formula [VIII], where R^{77} represents a

hydrocarbon group, especially a saturated or unsaturated hydrocarbon group having 1 to 4 carbon atoms.

[0077]

The present invention also provides, as a synthesis intermediate for the synthesis intermediate 1, a halogenated aryl compound represented by the formula [VII] (hereinafter called "the synthesis intermediate 2 of the present invention").

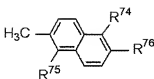
[0078]

The synthesis intermediate 2 of the present invention can be obtained by reacting a dimethylnaphtalene compound represented by the following formula [IX] with an N-halogenated succinimide represented by the following formula [X] under irradiation of light. For example, they can be reacted at 20 to 120°C under normal pressure for 30 to 48 hours in a solvent such as carbon tetrachloride, chloroform, benzene or chlorobenzene by using a light source such as a high-pressure mercury lamp, low-pressure mercury lamp, xenon lamp, halogen lamp, sunlight or fluorescent lamp.

[0079]

[Chemical Formula 64]

Formula [IX]



in the Formula [IX], where R^{74} and R^{75} may be the same or different, at least one of R^{74} and R^{75} represents a hydrogen atom, a cyano group, a nitro group, a trifluoromethyl group or a halogen atom, and R^{76} represents a hydrogen atom, a saturated or unsaturated alkyl group having 1 to 6 carbon atoms, or a substituted or unsubstituted aryl group.

[0080]

[Chemical Formula 65]

Formula [X]



in the Formula [X], where X represents a halogen atom.

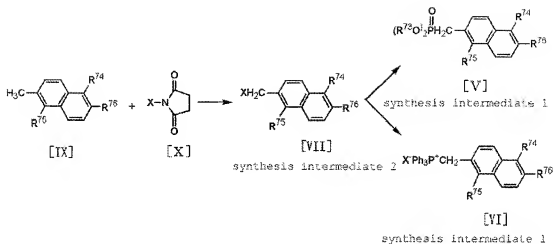
[0081]

The reactions for obtaining the above-mentioned, respective synthesis intermediates 1 and 2 can be expressed, for example, by the following reaction scheme 2.

[0082]

[Chemical Formula 66]

Reaction Scheme 2



[0083]

FIG. 1 to FIG. 4 show organic electroluminescent devices according to various embodiments of the present invention, respectively.

[0084]

FIG. 1 shows a transmission-type organic electroluminescent device A, in which emitted light 20 transmits through an anode 2 and light reflected by a cathode 3 is also obtained as emitted light 20. FIG. 2 illustrates a reflection-type organic electroluminescent device B, in which light reflected by an anode 2 is also obtained as emitted light 20 through a thin cathode 3 and the emitted light 20 can be observed from the side of a protective layer 4.

[0085]

In each of these drawings, numeral 1 indicates a substrate for forming the organic electroluminescent device. Glass, plastics or any other appropriate material is usable. When the organic electroluminescent device is used in combination with another display device, these devices can use a single substrate in common. In FIG. 1, the anode 2 is a transparent electrode and ITO, SnO_2 or the like can be used.

[0086]

In each of FIG. 1 and FIG. 2, an organic luminescent layer 5 contains the above-described aminostyrylnaphthalene compound of the present invention as a light emitting material. As a layer structure of the luminescent layer 5 for obtaining the organic electroluminescent 20, various known structures can be used. As will be described subsequently herein, when a material forming either a hole transport layer or an electron transport layer has light emitting property, for example, a structure with these transport layers stacked as thin films one over the other can be used.

[0087]

Further, the present invention does not prevent one or both of a hole transport layer and an electron

transport layer from using a structure, which is formed of thin films of plural materials stacked one over another, or a thin film of a composition, which has been obtained by mixing plural materials, to improve the performance of electron transportation within a range in which the objects of the present invention are satisfied. To improve the light emitting performance, at least one fluorescent material may also be used in such a structure as having a thin film of the fluorescent material held between a hole transport layer and an electron transport layer or in such a structure as having the fluorescent material incorporated in a hole transport layer or an electron transport layer or in both of them. In these cases, a thin film for controlling the transport of holes or electrons may also be included in the layer structure.

[0088]

As the aminostyrylnaphthalene compound represented by the formula [I], [II] or [III] is equipped with both of electron transporting ability and hole transporting ability, the compound can be used as a mixed luminescent layer together with an electron transporting material or as a mixed luminescent layer together with a hole transporting material in the device structure. Further, a mixed layer with the compound contained

therein can be used as a light emitting material in such a structure as having the mixed layer held between an electron transport layer and a hole transport layer.

[0089]

FIG. 1 and FIG. 2 each depicts the cathode 3. As an electrode material, it is possible to use an alloy or stacked structure of an active metal such as Li, Mg or Ca with a metal such as Ag, Al or In. In a transmission-type organic electroluminescent device, a light reflectance suited to its application purpose can be obtained by adjusting the thickness of its cathode. In each of FIG. 1 and FIG. 2, the protective film 4 has sealing property, and its effect can be enhanced by forming it into such a structure as covering the organic electroluminescent device in its entirety. Any suitable material can be used insofar as air tightness can be maintained.

[0090]

In each organic electroluminescent device according to the present invention, the organic layer may have an organic layer structure (single heterostructure) with a hole transport layer and an electron transport layer stacked one over the other, and as the material forming the hole transport layer or the electron

transport layer, a mixed layer with the aminostyrylnaphthalene compound contained therein can be used. As an alternative, the organic layer may have an organic layer structure (double heterostructure) with a hole transport layer, a luminescent layer and an electron transport layer stacked one over another, and as the material forming the luminescent layer, a mixed layer with the aminostyrylnaphthalene compound contained therein can be used.

[0091]

Other embodiments of organic electroluminescent devices each having such an organic layer structure will hereinafter be described. FIG. 3 illustrates an organic electroluminescent device C of the single heterostructure, which has a layer structure formed of a light-transmitting anode 2, an organic layer 5a, and a cathode 3 stacked one over another on a light-transmitting substrate 1. The organic layer 5a is composed of a hole transport layer 6 and an electron transport layer 7. The layer structure is sealed with a protective layer 4.

[0092]

In the case of a layer construction with a luminescent layer omitted as shown in FIG. 3, emitted light 20 of a predetermined wavelength is produced from

an interface between the hole transport layer 6 and the electron transport layer 7. The emitted light can be observed from the side of the substrate 1.

[0093]

FIG. 4, on the other hand, depicts an organic electroluminescent device D of the double heterostructure, which has a layer structure formed of a light-transmitting anode 2, an organic layer 5b, and a cathode 3 stacked one over another on a light-transmitting substrate 1. The organic layer 5b is composed of a hole transport layer 10, a 11 and an electron transport layer 12. The layer structure is sealed with a protective layer 4.

[0094]

When a DC voltage is impressed across the anode 2 and the cathode 3 in the organic electroluminescent device depicted in FIG. 4, holes injected from the anode 2 and electrons injected from the cathode 3 are caused to reach the luminescent layer 11 through the hole transport layer 10 and the electron transport layer 12, respectively. As a result, recombination of electrons and holes takes place in the luminescent layer 11 to form singlet excitons, from which emission of light of a predetermined wavelength is produced.

[0095]

In each of the above-mentioned organic electroluminescent devices C, D, a light-transmitting material such as glass or plastics can be used as the substrate 1 as desired. When the organic electroluminescent device is used in combination with another display device or when a plurality of layer structures as shown in FIG. 3 or FIG. 4 are arranged in the form of a matrix, for example, the substrate may be used in common. Further, the devices C and D can each take either a transmission-type structure or a reflection-type structure.

[0096]

The anode 2 is a transparent electrode, and ITO (indium tin oxide), SnO_2 or the like can be used. Between the anode 2 and the hole transport layer 6 (or the hole transport layer 10), a thin film made of an organic material or an organometallic compound may be arranged to improve the charge injection efficiency. When the protective layer 4 is formed of a conductive material such as a metal, the anode 2 may be provided on side walls thereof with insulating films, respectively.

[0097]

In the organic electroluminescent device C, the

organic layer 5a is formed of the hole transport layer 6 and electron transport layer 7 stacked together. The above-described aminostyrylnaphthalene compound may be incorporated in one or both of these transport layers to convert them into a luminescent, hole transport layer 6 and/or a luminescent, electron transport layer 7. In the organic electroluminescent device D, on the other hand, the organic layer 5b is formed of the hole transport layer 10, the luminescent layer 11 with the above-described aminostyrylnaphthalene compound contained therein, and the electron transport layer 12, all of which are stacked together. In addition to this stacked structure, the organic layer 5b can take a variety of other stacked structures. For example, one or both of the hole transport layer and electron transport layer may be modified into luminescent transport layer or layers.

[0098]

Further, each hole transport layer may be formed of plural hole transporting materials stacked in layers one over another such that the hole transport layer is provided with improved hole transporting ability.

[0099]

In the organic electroluminescent device C, the luminescent layer may be the luminescent, electron

transport layer 7. Depending on a voltage impressed from a power supply 8, however, light may be emitted at the hole transport layer 6 or at its interfaces. In the organic electroluminescent device D, the luminescent layer may also be either the electron transport 12 or the hole transport layer 10 in addition to the layer 11. To improve the light emitting performance, however, it is desired to adopt such a structure that a luminescent layer 11, which makes use of at least one fluorescent material, is held between a hole transport layer and an electron transport layer. As an alternative, the organic layer may be constructed into such a structure that the fluorescent material is incorporated in one or both of the hole transport layer and the electron transport layer. In each of these structures, it is possible to include a thin film, which can serve to control the transport of holes or electrons (for example, a hole blocking layer, exciton generating layer or the like), in the layer structure to improve its light emitting efficiency.

[0100]

As a material for each cathode 3, an alloy of an active metal such as Li, Mg or Ca and a metal such as Ag, Al or In can be used. As an alternative, these metals may be used in a stacked structure. By selecting the

thickness and material of the cathode as desired, an organic electroluminescent device suited for a specific application can be fabricated.

[0101]

Each protective layer 4 acts as a sealing film. By providing the protective layer in such a structure as covering the organic electroluminescent device in its entirety, the charge injection efficiency and light emitting efficiency can be improved. Insofar as the organic electroluminescent device can be maintained airtight, any desired material can be selected for the protective layer 4, including single metals and alloys of aluminum, gold, chromium and the like.

[0102]

A current to be impressed to each of the above-described organic electroluminescent devices is generally a direct current, but instead, a pulsed current or an alternating current may be used. Neither the current value nor the voltage value are particularly limited insofar they fall within such ranges that the device would remain undamaged. In view of the power consumption and service life of the organic electroluminescent device, however, it is desired to make the organic electroluminescent device efficiently emit light with as

smaller electric energy as possible.

[0103]

FIG. 5 shows an illustrative construction of a flat display making use of an organic electroluminescent device according to the present invention. For example, in the case of a full-color display as depicted in the drawing, an organic layer 5 (5a, 5b) which can emit primaries of red (R), green (G) and blue (B) are arranged between cathodes 3 and anodes 2. The cathodes 3 and anodes 2 can be arranged in the form of stripes crossing each other. By a luminance signal circuit 14 and a control circuit 15 equipped with a built-in register, signal voltages are selectively impressed to the cathodes 3 and anodes 2 such that light is emitted from the organic layers at positions (pixels) where the selected cathodes 3 and anodes 2 intersect with each other in a single matrix system or active matrix system.

[0104]

Described specifically, FIG. 5 shows by way of example an 8×3 RGB single matrix, in which stacked layers 5 each formed of a hole transport layer and at least one of a luminescent layer and electron transport layer are arranged between the cathodes 3 and the anodes 2 (see FIG. 3 or FIG. 4). The cathodes and anodes are

constructed such that they are patterned in the form of stripes and are arranged at right angles relative to each other in the form of a matrix and, by the control circuits 15 and 14 equipped with the built-in shift register, signal voltages are impressed in time sequence to emit light at their crossing positions. EL devices of such a construction can also be used as picture reproducers, to say nothing of displays for characters, signs and the like. Further, the striped patterns of the cathodes 3 and anodes 2 can be arranged for each of red (R), green (G) and blue (B) to construct a multicolor or full-color, full solid-state flat panel display.

[0105]

[Examples]

Certain examples of the present invention will next be described, although the present invention shall not be limited to them.

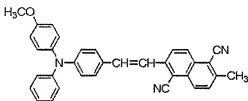
[0106]

Example 1

A transmission-type organic electroluminescent device of the single heterostructure was fabricated by using the compound of the above-described structural formula (20)-35 as a luminescent, electron transport layer and 4,4'-bis[N,N'-di(1-naphthyl)-N,N'-

diphenyl]biphenyldiamine (α -NPD) of the below-described structural formula as a hole transport layer.

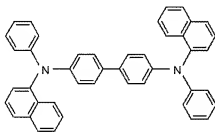
Structural formula (20)-35



[0107]

[Chemical Formula 67]

α -NPD



[0108]

Firstly, a 30 mm \times 30 mm glass substrate with a 100-nm thick ITO anode formed on a surface thereof was set in a vacuum evaporation system. A metal mask having a plurality of 2.0 mm \times 2.0 mm unit openings formed therethrough was disposed as an evaporation mask in a proximity of the substrate, and by vacuum evaporation, α -NPD as a hole transporting material was deposited to a thickness of 140 nm under a vacuum of 10^{-4} Pa or lower. Further, the compound of the above structural formula

(20)-35 was deposited as a luminescent, electron transporting material to a thickness of 55 nm in contact with the hole transport layer. The deposition rate was set at 0.2 nm/sec in each of the above vacuum evaporation steps.

[0109]

As cathode materials, Mg and Ag were adopted. By vacuum evaporation, they were also deposited at deposition rates of 1 nm/sec to thicknesses of 50 nm (Mg film) and 150 nm (Ag film), respectively, to form a stacked film of Mg and Ag, so that an organic electroluminescent device according to the Example 1 was fabricated as illustrated in FIG. 3.

[0110]

To the organic electroluminescent device fabricated as described above, a forward bias d.c. voltage was applied in a nitrogen atmosphere to evaluate its light emitting characteristics. Emitted light had a red color. As a result of spectrometry, a spectrum having an emission peak around 610 nm was obtained. The electroluminescent spectrum is shown in FIG. 6. In the spectrometry, a spectroscopy manufactured by OTSUKA ELECTRONICS CO., LTD. and equipped with a photodiode array as a detector was used. In addition, a voltage-

luminescence measurement was conducted. As depicted in FIG. 7, a luminance of 490 cd/m² was obtained at 8 V.

[0111]

After the fabrication, the organic electroluminescent device was left over for a month under a nitrogen atmosphere, but no degradation was observed on the device. Further, the organic electroluminescent device was subjected to forced degradation at an initial luminance of 100 cd/m² by applying a current at a constant value to continuously emit light. It took 800 hours until the luminance dropped to half.

[0112]

Example 2

A transmission-type organic electroluminescent device of the double heterostructure was fabricated by using the above-described α -NPD as a hole transport layer and the compound of the above-described structural formula (20)-35 as a luminescent layer.

[0113]

Firstly, a 30 mm \times 30 mm glass substrate with a 100-nm thick ITO anode formed on a surface thereof was set in a vacuum evaporation system. A metal mask having a plurality of 2.0 mm \times 2.0 mm unit openings formed therethrough was disposed as an evaporation mask in a

proximity of the substrate, and by vacuum evaporation, α -NPD as a hole transporting material was deposited to a thickness of 140 nm under a vacuum of 10^{-4} Pa or lower. Further, the compound of the above structural formula (20)-35 was deposited as a light emitting material to a thickness of 40 nm in contact with the hole transport layer. The deposition rate was set at 0.2 nm/sec in each of the above vacuum evaporation steps. As an electron transport layer, tris(8-quinolinol)aluminum (Alq_3) of the below-described structural formula was deposited further in contact with the luminescent layer. The electron transport layer made of Alq_3 was deposited to a thickness of 50 nm. The deposition rate was set at 0.2 nm/sec.

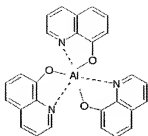
[0114]

As cathode materials, Mg and Ag were adopted. By vacuum evaporation, they were also deposited at deposition rates of 1 nm/sec to thicknesses of 50 nm (Mg film) and 150 nm (Ag film), respectively, to form a stacked film of Mg and Ag, so that an organic electroluminescent device according to the Example 2 was fabricated as illustrated in FIG. 4.

[0115]

[Chemical Formula 68]

Alq_3



[0116]

To the organic electroluminescent device fabricated as described above, a forward bias d.c. voltage was applied in a nitrogen atmosphere to evaluate its light emitting characteristics. Emitted light had a red color. As a result of spectrometry, an emission spectrum similar to that obtained in Example 1 was obtained. In the spectrometry, a spectroscopy manufactured by OTSUKA ELECTRONICS CO., LTD. and equipped with a photodiode array as a detector was used. In addition, a voltage-luminescence measurement was conducted. As depicted in FIG. 7, a high luminance of 1150 cd/m² was obtained at 8 V.

[0117]

After the fabrication, the organic electroluminescent device was left over for a month under a nitrogen atmosphere, but no degradation was observed on the device. Further, the organic electroluminescent device was subjected to forced degradation at an initial luminance of 100 cd/m² by applying a current at a constant

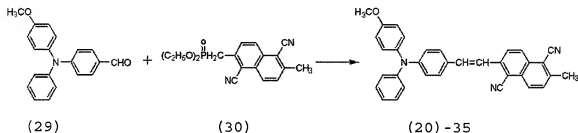
value to continuously emit light. It took 1300 hours until the luminance dropped to half.

[0118]

Example 3

<Synthesis Example of (Aminostyryl)naphthalene

Compound (Structural Formula (20)-35)>



[0119]

Under a nitrogen atmosphere, a Wittig-Horner reagent (30) (7.00 g, 20.3 mmol) was suspended in methanol (100 mL) over an ice bath. Under stirring, sodium methoxide (1.20 g, 22.2 mmol) was added little by little, followed by stirring for 30 minutes. 4-[N,N'-(4-methyldiphenyl)]aminobenzaldehyde (29) (5.60 g, 18.5 mmol) was added in two portions, and the temperature of the resulting mixture was allowed to rise, as was, from 0°C to room temperature, at which the mixture was stirred for six hours.

[0120]

The resulting precipitate was collected by filtration, and then recrystallized three times from

tetrahydrofuran (THF)-toluene-methanol to afford an orange-color powder (3.05 g). The product was identified to be the target compound by ^1H NMR and FAB-MS analyses (isolation yield: 30%). Its identification data were as described below, and its ^1H NMR spectrum is presented in FIG. 8.

[0121]

^1H NMR (400 MHz, CDCl_3) δ (ppm): 2.77(s, 3H), 3.75(s, 3H), 6.88(d, 2H), 7.00-7.13(m, 7H), 7.27-7.53(m, 6H), 7.59(d, 1H), 8.04(d, 1H), 8.31(d, 2H).

[0122]

Incidentally, a solution of the product in THF had a visible absorption maximum at 437 nm and a fluorescence maximum wavelength at 612 nm. The relative fluorescence quantum yield of the product in dioxane was 0.97, that is, very high.

[0123]

[Action Effect of the Invention]

According to the present invention, the aminostyrylnaphthalene compound represented by the formula [I], [II] or [III] is excellent in light emitting properties, and moreover, shows amorphous properties advantageous for film formability by vacuum deposition or the like and also durability. The use of the

aminostyrylnaphthalene compound makes it possible to provide an organic electroluminescent device capable of producing high-luminance and stable emission of red light at an optimal wavelength.

[Brief Description of the Drawings]

[FIG. 1]

FIG. 1 is a schematic fragmentary cross-sectional view of an organic electroluminescent device according to an embodiment of the present invention.

[FIG. 2]

FIG. 2 is a schematic fragmentary cross-sectional view of an organic electroluminescent device according to another embodiment of the present invention.

[FIG. 3]

FIG. 3 is a schematic fragmentary cross-sectional view of an organic electroluminescent device according to a further embodiment of the present invention.

[FIG. 4]

FIG. 4 is a schematic fragmentary cross-sectional view of an organic electroluminescent device according to a still further embodiment of the present invention.

[FIG. 5]

FIG. 5 is a construction diagram of a full-color

flat display making use of an organic electroluminescent device according to the present invention.

[FIG. 6]

FIG. 6 is an emission spectrum diagram of an organic electroluminescent device according to an Example of the present invention.

[FIG. 7]

FIG. 7 is a voltage-luminance characteristic diagram of the organic electroluminescent device according to an Example of the present invention.

[FIG. 8]

FIG. 8 is a ^1H NMR spectrum diagram of an aminostyrylnaphthalene compound suited for use in an organic electroluminescent device according to an Example of the present invention.

[Description of Reference Numerals]

1 ... substrate, 2 ... transparent electrode or anode, 3 ... cathode, 4 ... protective layer, 5, 5a, 5b ... organic layer, 6 ... hole transport layer, 7 ... electron transport layer, 8 ... power supply, 10 ... hole transport layer, 11 ... luminescent layer, 12 ... electron transport layer, 14 ... luminance signal circuit 14 and a control circuit, 15 ... control circuit, 20 ... emitted light, A, B, C, D ... organic electroluminescent device.

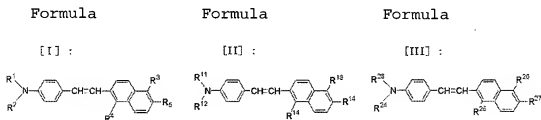
[Name of Document] Abstract of the Disclosure

[Abstract]

[Object] To provide an organic electroluminescent device using a compound suitable as an organic luminescent material capable of producing high-luminance and stable emission of red light at an optimal wavelength, the compounds, synthesis intermediates thereof, and production processes of the same.

[Solving Means] An organic electroluminescent device includes an organic layer having a light-emitting area and arranged between an anode and a cathode, and an organic material which emits light when a current is supplied. The organic layer contains in at least a part thereof at least one aminostyrylnaphthalene compound represented by the following formula [I], [II] or [III]:

[Chemical Formula 1]



wherein R^1 , R^2 , R^{11} , R^{12} , R^{23} and R^{24} are each a phenyl or naphthyl group, R^3 , R^4 , R^{13} , R^{14} , R^{25} and R^{26} are each an electron attracting group such as a cyano group, and R^5 , R^{15} and R^{27} are each a substituent group such as an

alkyl group.

[Selected Drawing] FIG. 6

[Name of Document] Drawings

[FIG. 1]

A: Transmission-type Organic Electroluminescent Device

- 1: Substrate
- 2: Transparent Anode
- 3: Cathode
- 4: Protective Layer
- 5: Luminescent Layer

[FIG. 2]

B: Reflection-type Organic Electroluminescent Device

- 1: Substrate
- 2: Anode
- 3: Thin Cathode
- 4: Protective Layer
- 5: Luminescent Layer

[FIG. 3]

C: Organic Electroluminescent Device

- 1: Substrate
- 2: Anode
- 3: Cathode
- 4: Protective Layer
- 6: Hole Transport Layer

7: Electron Transport Layer

[FIG. 4]

D: Organic Electroluminescent Device

1: Substrate

2: Anode

3: Cathode

4: Protective Layer

10: Hole Transport Layer

11: Luminescent Layer

12: Electron Transport Layer

[FIG. 5]

14: Luminance Signal Circuit

15: Control Circuit

[FIG. 6]

A: Intensity (Unit: Optional)

B: Wavelength (nm)

[FIG. 7]

A: Luminance (cd/m^2)

B: Voltage (V)

C: Example 1

D: Example 2

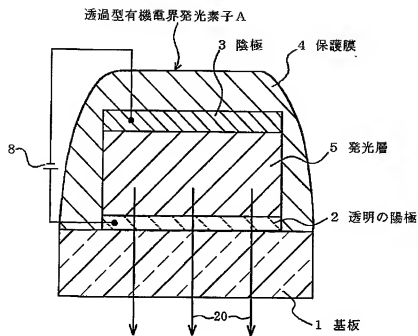
[FIG. 8]

A: TMS (Tetramethylsilane)

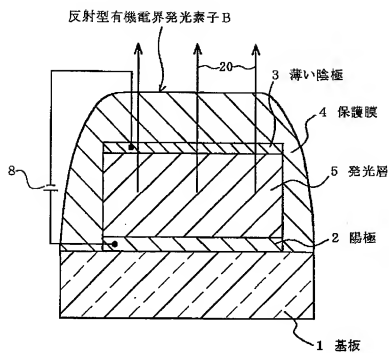
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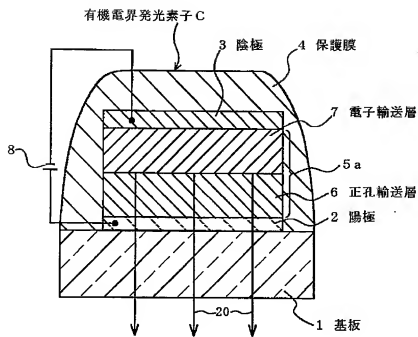
【図1】



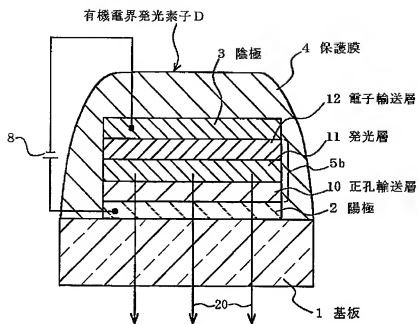
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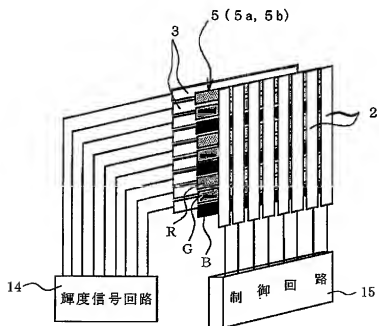
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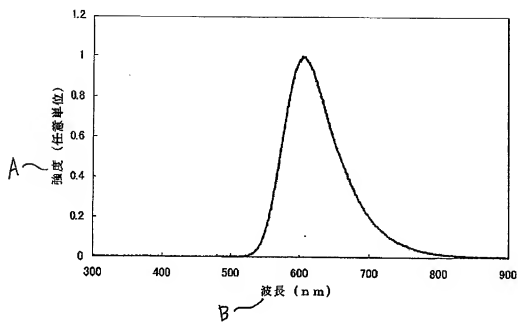
【圖4】



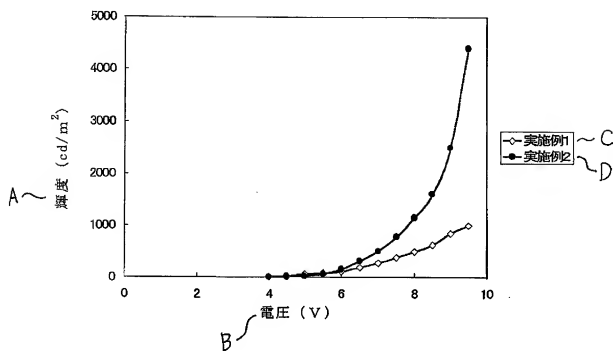
【圖5】



【図6】



【图7】



【図8】

